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Research Front: Tellurium in Biological and Environmental Systems: after Fukushima

Losses and environmental aspects of a byproduct metal: A review of tellurium

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Environmental context. Studies involving modelling are increasingly being performed to better understand how technology-critical elements such as tellurium are transported and accumulated in man-made technological systems. The resulting 'anthropogenic cycles' provide estimates of current and anticipated future material releases to the environment, and their associated environmental implications. This information complements data on natural cycles in which the subsequent transport and fate of tellurium in the environment can be examined.

Abstract. Global demand for tellurium has greatly increased owing to its use in solar photovoltaics. Elevated levels of tellurium in the environment are now observed. Quantifying the losses from human usage into the environment requires a life-cycle wide examination of the anthropogenic tellurium cycle (in analogy to natural element cycles). Reviewing the current literature shows that tellurium losses to the environment might occur predominantly as mine tailings, in gas and dust and slag during processing, manufacturing losses, and in-use dissipation (situation in around 2010). Large amounts of cadmium telluride will become available by 2040 as photovoltaic modules currently in-use reach their end-of-life. This requires proper end-of-life management approaches to avoid dissipation to the environment. Because tellurium occurs together with other toxic metals, e.g. in the anode slime collected during copper production, examining the life-cycle wide environmental implication of tellurium production requires consideration of the various substances present in the feedstock as well as the energy and material requirements during production. Understanding the flows and stock dynamics of tellurium in the anthroposphere can inform environmental chemistry about current and future tellurium releases to the environment, and help to manage the element more wisely.

Keywords: Material flow analysis, industrial ecology, anthropogenic metal cycles,

1. Introduction

Tellurium is a very rare element with a crustal abundance of only 3 parts per billion in Earth's upper crust (Goldfarb et al. 2017). Globally, tellurium is used in photoreceptors for the production of solar cells (40% of global consumption), thermoelectric production (30%), metallurgy (15%), rubber applications (5%), and other uses (10%) (USGS 2018). In recent years, concerns have been raised over the environmental and human health issues related to some forms of tellurium in the environment which can be highly toxic (Taylor 1996). For

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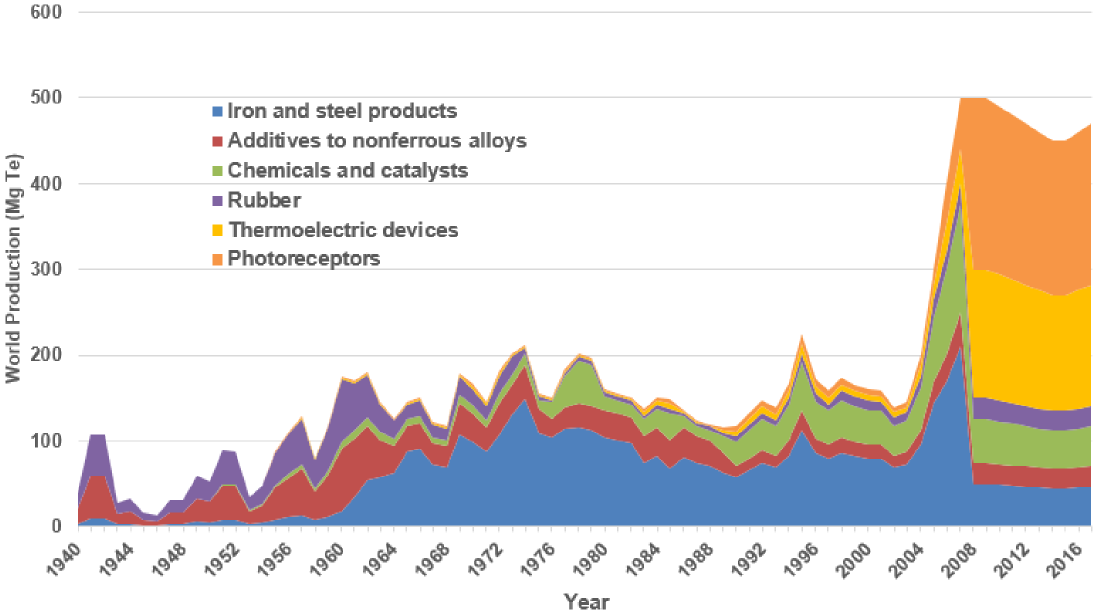
example, tellurium is frequently released into the environment as a byproduct during metal production in the smelting stage, as well as from the mining and burning of coal and oil. Recent work has found that anthropogenic activities have significantly increased atmospheric tellurium levels in the environment (Wiklund et al. 2018).

Globally, tellurium production has increased almost five-fold from around 100 megagrams (Mg) in 1940 to approximately 470 Mg in 2017 (Kavlak and Graedel 2013; USGS 2018). Recent years have witnessed an increasing demand especially for cadmium-telluride photovoltaic cells and thermoelectric devices (Figure 1).

Figure 1. World production of tellurium from 1940 to 2017 divided into the six principal end uses[a] (Megagrams (Mg)).



[a] Data from 1940 to 2010 based on (Kavlak and Graedel 2013). (Goldfarb et al. 2017) report a global production of 450 Mg tellurium in 2014 and a linear decrease from 2010 to 2014 was assumed. Data for years 2016 and 2017 are based on (USGS 2018) adding 50 Mg tellurium to account for the (information withheld) refinery production of the United States (Kavlak and Graedel 2013).

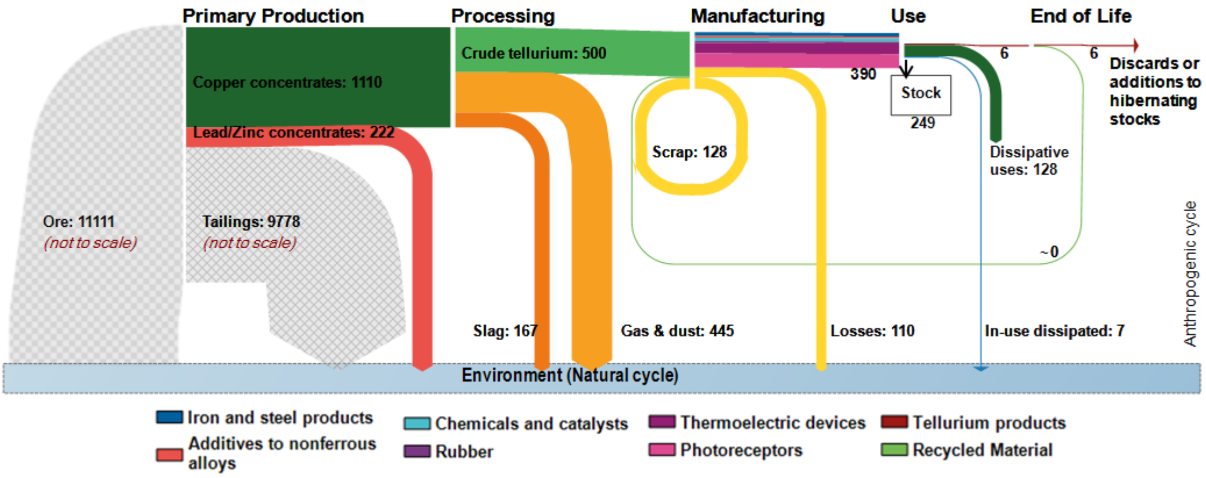
Scenario studies show that the demand for tellurium from cadmium-telluride cells alone could result in future demands of above 2,500 Mg by 2050 (Elshkaki and Graedel 2013). Recent research also highlights that if cadmium telluride photovoltaics account for more than 3% of electricity generation by 2030, the required growth rates for the production of tellurium might exceed historically-observed production growth rates of the element (Kavlak et al. 2015). According to Kavlak and colleagues, required annual tellurium production in 2030 could also exceed tellurium reserves (Kavlak et al. 2015) (current reserves are estimated at about 31,000 Mg tellurium (USGS 2018). However, the authors note that reserve estimates are constantly revised to reflect newly identified mineable deposits. While renewable energy technologies such as thin-film photovoltaics have the potential to contribute to climate change mitigation, there are also concerns over the future availability of tellurium. For example, about half of the criticality studies which examine materials on the basis of possible supply shortages and their economic importance find that tellurium is indeed a critical element (Hayes and McCullough 2018).

As demands for the element are on the rise and increasing quantities of tellurium-containing products are present in society, it is important to define and quantify the anthropogenic rates of supply and demand including possible transformation losses into the environment. Such information is relevant to environmental chemists studying the subsequent transport and fate of tellurium in the environment. While the characterization of elemental cycles has a rich history in environmental chemistry, characterizing the anthropogenic life cycles of both substances and goods through material flow analysis (MFA) has developed more recently (Brunner and Rechberger 2016; Müller et al. 2014). MFA can provide information on the material stocks in the anthroposphere (in-use stocks) and in nature (e.g., in soils, tailings, and mining wastes), and estimate anticipated emissions in the future when products reach their end-of-life, thus complementing natural cycles (Klee and Graedel 2004; Sen and Peucker-Ehrenbrink 2012; Nuss and Blengini 2018). In the policy context, MFA provides an important foundation, e.g., for resource efficiency, raw materials, and circular economy policies (EC 2018; BMUB 2016; EC 2008).

2. Anthropogenic tellurium cycle

A literature review examining the scientific and grey literature on anthropogenic tellurium cycles was undertaken (Table S1 in the appendix). Linkages to natural cycles exist when tellurium is extracted together with copper or other host metals from the lithosphere or when losses during the metal's life-cycle to the environment (e.g., as tailings, slag, gas & dust, manufacturing losses, or in-use dissipation) take place. Based on research previously undertaken by (Kavlak and Graedel 2013) and others, data are rearranged and a Sankey visualization of the global anthropogenic tellurium cycle around 2010 is generated which highlights the magnitude of tellurium flows (width of the arrows), in-use stocks (i.e., tellurium in long-living products), and possible losses from the anthroposphere to the environment (i.e. if tellurium losses are not addressed through proper mine-, materials- and waste-management) (Figure 2). Please note that the data presented in Figure 2 represent informed estimates rather than highly certain values due to data limitations and uncertainties in the existing data and estimates provided in the literature (see also (Kavlak and Graedel 2013) for further details).

Figure 2. Global anthropogenic tellurium cycle around 2010 including losses to the environment[a]. All number in megagrams (Mg) tellurium.



[a] Own compilation based on data by (Kavlak and Graedel 2013; Nassar et al. 2012), end-uses by (USGS 2018), and the fraction of tellurium in-use dissipated by (Ciacci et al. 2015). Only tellurium flows associated with copper production processes are quantified. The fraction of in-use dissipation is based on (Ciacci et al. 2015). Note that the flow magnitudes of ores and tailings are several orders of magnitude larger than for other flows, hence, these are not shown to scale.

Primary production and processing:

Tellurium is presently recovered mostly as a byproduct of the mining of porphyry copper deposits and there are only two deposits in the world (located in China and Sweden) from which tellurium is obtained as a primary ore. However, these account only for about 15 percent of the annual global production of tellurium (Goldfarb et al. 2017). The principal source of tellurium (~90% in 2017) is anode sludge produced during the electrolytic copper refining, and the remainder is produced from skimmings at lead refineries and from flue dusts and gases during the smelting of copper, lead-zinc ores, and bismuth (USGS 2018, 2016). In 2017, tellurium was produced mainly in China (280 metric tons (MT) of tellurium content), the United States (50 MT)[b], Japan (38 MT), Sweden (40 MT), Russia (35 MT), Canada (20 MT), and Bulgaria (4 MT) (USGS 2018; BGS 2017). Only small amounts of tellurium (< 1%) were from secondary sources, e.g., from scrapped selenium-tellurium photoreceptors used in paper copiers in Europe, while recycling from cadmium telluride (CdTe) solar cells is currently limited due to the fact that most solar cells are relatively new and have not yet reached their end-of-life (Graedel et al. 2011; USGS 2018). Tellurium reserves are predominantly located in China, Peru, and the United States (USGS 2018).

Around 90% of the tellurium is lost to tailings during mining and concentration and 55% to slag, gas and dust during the processing step consisting of smelting and anode slime treatment (Figure 2) (Kavlak and Graedel 2013). The anode slime collected during copper production contains tellurium as well as various other metals (e.g., bismuth, antimony, selenium, lead, gold, silver, arsenic, and nickel) (Knockaert 2011). While the effective management of mine tailings reduces the loss of tellurium and other metals into the environment (Reid et al. 2009), geochemical weathering, e.g., of unreclaimed mine wastes can result in the release of bioaccessable tellurium into the environment (Bullock et al. 2017; Qin et al. 2017). Furthermore, recent work has found that near copper smelting operations in Canada the tellurium concentrations in lakes increased over 100 times after opening of the smelter in 1930 (Wiklund et al. 2018). Enriched tellurium concentrations in sediment cores have also been attributed to copper processing in other parts of North America (Dolor et al. 2009) Given that the primary production and subsequent processing steps result in the largest amount of tellurium losses to the environment (Figure 2), it is important that more empirical studies on tellurium releases and the possible further cycling in the environment are conducted.

Manufacturing and use:

The fabrication and manufacturing stage starts with crude tellurium (copper telluride) from the anode slimes of copper refining (Hoffmann et al. 2011). The overall loss rate of converting copper telluride into downstream products is around 10% for end-uses other than electronics (Kavlak and Graedel 2013). Producing commercial grade tellurium used in electronics with purities of 4N (99.99%) or 5N (99.999%) results in additional losses. Tellurium finds use in the manufacture of a variety of products including iron and steel products, additives to nonferrous alloys, chemicals and catalysts, rubber, thermoelectric devices, and photoreceptors (USGS 2018). Of these end-uses, tellurium can currently not be made available for functional reuse from metallurgical additives, chemicals, and rubber products (termed “dissipative uses” in Figure 2). However, inherently dissipative uses in which tellurium is lost into the environment (termed: “in-use dissipated” in Figure 2) include its application as a germicide and fungicide, as a lubricant and grease in electronics (organic tellurides), as a jelling promoter in explosives (sodium tellurite), and in medical and biological uses (e.g., organic tellurobromides and terpene ether tellurocyanates) (Hoffmann et al. 2011; Ciacci et al. 2015). Depending on the chemical form dissipated and its bioavailability, uptake and further natural cycling will vary (see other papers in this special issue).

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[b] The refinery production of the USA is withheld by (USGS 2018) and an estimate for 2016 used from (BGS 2017).

While the life-times of the dissipative end-uses are short (in the MFA model they are assumed to be less than 1 year), tellurium-containing products such as thermoelectric devices and photoreceptors have assumed life-times in the MFA model of approximately 10 years and 30 years, respectively. Figure 2 also highlights that the in-use stock of tellurium is growing (the net change in in-use stocks equaled 249 Mg in 2010). Hence, large amounts of tellurium from electronics (especially CdTe photovoltaics currently in use) will only reach their end-of-life in the future. For example, Marwede and colleagues estimate that tellurium recycled from end-of-life (EoL) photovoltaic modules in a single year could make up around 40-50% of tellurium demand for photovoltaics by 2040, or even surpass feedstock needs by 40 metric tons (Marwede and Reller 2012). This is important information for environmental chemists because increasing amounts of CdTe waste feedstock, if not properly managed, might result in losses to the environment. Concerns over the toxicity of the feedstock materials (CdTe) exist although the health hazards presented by cadmium and tellurium vary as a function of the compounds specific toxicity, its physical state, and the mode of exposure, and they have not been fully examined yet (Fthenakis 2018). In order to avoid losses of CdTe feedstocks into the environment as increasing amounts of CdTe become available at end-of-life in the future, material efficiency must be substantially improved and collection and recycling systems have to be built up. For this purpose, a number of directives have been drafted to increase the collection and recycling of photovoltaic panels (Padoan et al. 2019). They include the EU Waste Electrical and Electronic Equipment Directive 2012/19/EU which aims at the recovery and preparation for re-use and recycling of photovoltaic panels to a percentage of 85% and 80%, respectively starting from 15 August 2018 (EU 2012). Other legislation not directly linked to the recycling of photovoltaic panels include EU legislation restricting the use of hazardous substances in electrical and electronic equipment (RoHS Directive 2002/95/EC and updated Directive 2011/65/EU) (EU 2011). It requires heavy metals such as cadmium and flame retardants to be substituted by safer alternatives. China also implemented a RoHS directive following the EU example (Padoan et al. 2019). Finally, the PV cycle association founded in 2007 aims to set up voluntary take-back and recycling schemes for EoL photovoltaic modules (PV CYCLE 2019).

Waste Management:
Current average end-of-life recycling rates for tellurium (reflecting the total material input into the production system originating from the recycling of post-consumer scrap) are currently only around 1% (Talens Peirò et al. 2018; Graedel et al. 2011). Recent work by (Ciacci et al. 2015) estimates that potential tellurium recycling could in theory increase to about 85% assuming almost complete recycling across the element’s various end-uses but considering that tellurium cannot be recycled from its uses as a vulcanizing agent and accelerator in the processing of rubber (about 5% of global use) and other dissipative uses (e.g., in lubricants and greases in electronics, in explosives, or medical and biological uses) equaling about 10% of global use. However, current inputs of secondary materials are limited by the fact that in-use stocks are still growing and that functional reuse for many end-uses is simply not possible with current collection and recycling systems.

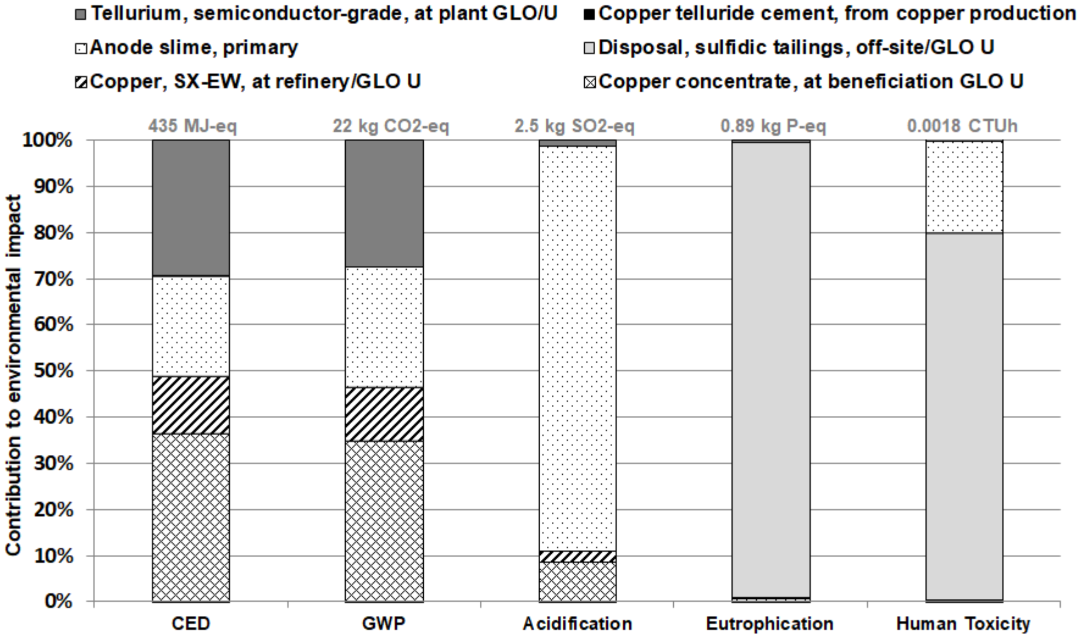
3. Environmental Implications

Environmental and human health issues can occur directly as a result of losses into the environment of some forms of tellurium which can be toxic (Taylor 1996). Furthermore, environmental impacts are also associated with the life-cycle wide energy and materials needs, emissions to air/soil/water, and waste outputs and treatment associated with the production of tellurium and subsequent manufactured products (occurring both on-site as well as up-and downstream the tellurium production processes). Because the anode slimes collected during copper production contain, besides tellurium, other potentially toxic elements such as bismuth, antimony, selenium, lead, gold, silver, arsenic, and nickel, dissipation of these into the environment can also lead to adverse environmental implications.

Figure 3 highlights the main process contributions to environmental impacts associated with the production of 1 kg of semiconductor grade tellurium using life-cycle assessment (LCA) (ISO 2006a, 2006b) (the higher grade tellurium was chosen due to data availability). Data are taken from (Nuss and Eckelman 2014) and are based on the Ecoinvent database v2.2 (ecoinvent 2010) using economic allocation to allocate environmental impacts between tellurium and the other metals obtained during the production route (e.g., silver, selenium, and copper). These estimates were publicly available and considered to provide a sufficient first overview of the relative contribution of different unit processes to environmental impacts associated with tellurium production. However, note that the estimates shown can vary from results obtained, e.g., using the most recent version of Ecoinvent (v3.5 at the time of writing) as background data such as regional energy mixes, transportation and waste treatment change over time (see www.ecoinvent.org), and results depend on the allocation approach chosen. Hence, for more in-depth examinations of tellurium environmental impacts it is recommended to consult the latest available life-cycle inventory datasets and carry out additional data collection.

Despite a number of data challenges for tellurium (e.g., data are only available for a limited number of production sites), LCA allows a first examination of various environmental impact categories at mid-point including, e.g., cumulative energy demand (CED), climate change (global warming potential (GWP)), acidification, eutrophication, and human toxicity associated with the production of tellurium metal.

Figure 3. Process contributions to cumulative energy demand (CED), global warming potential (GWP), terrestrial acidification, freshwater eutrophication, and human toxicity associated with the cradle-to-gate production of 1 kg of semiconductor grade tellurium[a].



[a] Data are taken from (Nuss and Eckelman 2014) and are originally based on the Ecoinvent 2.2 database (ecoinvent 2010) Environmental impact categories include: global warming potential (GWP) (IPCC 2007 GWP 100a v1.02 (Goedkoop et al. 2008)), cumulative energy demand (CED) (Cumulative Energy Demand v1.08 (Goedkoop et al. 2008)), terrestrial acidification, freshwater eutrophication, and human toxicity (USETox ® v1.02 with recommended and interim characterization factors (CTUh = Comparative Toxic Unit for human) (Rosenbaum et al. 2008)). Note that results might

differ from more recent versions of Ecoinvent as, e.g., background data such as energy mixes, transportation, waste treatment have changed. Impacts also depend on the allocation approach chosen and absolute numbers shown should only be considered as approximate estimates of potential environmental impacts. SX-EW stands for hydrometalurgically won copper.

Figure 3 highlights that the production of copper concentrate, anode slime from copper refining (containing tellurium and other metals), and tellurium metal result in the largest contributions to cumulative energy demand and global warming potential (two left-hand bars in Figure 3). This is largely due to the use of fossil fuels and electricity during the mining and refining stages of tellurium production. Acidification impacts are dominated by the emissions of sulfur dioxide and nitrogen oxides during the electrolytic copper refining where anode slime containing tellurium is obtained (center bar). For eutrophication and human toxicity (Rosenbaum et al. 2008), the disposal of sulfidic tailings results in the largest environmental burdens (two right-hand bars in Figure 3). For sulfidic tailings disposal the environmental and human health impacts are largely a result of potential leaching of metals such as manganese, arsenic, selenium, and others in groundwater to the environment during the disposal of sulfidic tailings (please note that only a generic model for sulfidic tailings disposal is used in this study based on (Doka 2008)). Refinery production leads to additional potential impacts associated with the release of byproduct metals to air as well as energy requirements and resulting emissions of carbon dioxide to air (contributing to climate change).

4. Conclusions

Tellurium is of increasing technological importance and elevated levels in the environment are now observed. Understanding possible losses of the element from human activities requires quantifying the stocks and flows in the anthroposphere (anthropogenic tellurium cycle). This can support environmental chemistry research in understanding the current and anticipated magnitude of tellurium flows to the environment, e.g., to develop monitoring approaches targeting specific life-cycle stages. Because tellurium occurs as a companion (byproduct) metal with copper and the feedstocks and subsequent tellurium products (e.g., cadmium telluride photovoltaics) contain other potentially toxic metals (e.g., arsenic, cadmium, selenium), it is important to also consider their potential environmental impacts as losses to the environment takes place (e.g., during the treatment of sulfidic tailings or losses to the air during the smelting stage). In the future, large quantities of tellurium will become available as photovoltaic modules reach their end-of-life and dissipation of cadmium telluride to the environment might occur if waste streams are not properly managed. Future research should aim to increasingly combine material flow analysis and natural cycle data to obtain more complete tellurium cycles and support the sustainable management of this scarce element.

5. Acknowledgements

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6. Conflict of Interest

The authors declare no conflicts of interest.

7. Appendix

Literature review:

The literature review performed in this study aimed at having a representative sample of studies from the literature that describe either the full or parts of the anthropogenic tellurium cycle, i.e., from mining to smelting to fabrication and manufacture, use, and waste management and recycling at end-of-life.

For this, a literature search was carried out using commonly used web search engines and academic interdisciplinary databases including Scopus and Google Scholar. The key words used in this search were "tellurium material flow" in the timeframe 2000-2018. The anchor title was complemented with other keywords including "anthropogenic cycle, material flow analysis, life-cycle assessment, anthropogenic cycle, criticality study, production". The aim was not to comprehensively cover the literature in the field, but to obtain a list of the most common and widely used sources and data for describing the anthropogenic flows and stocks of tellurium. The search includes global as well as regional and country studies.

Studies that focus on the host metal copper (Passarini et al. 2018; Ciacci et al. 2017; Graedel et al. 2002; Rechberger and Graedel 2002; Spatari et al. 2005; Chen and Graedel 2012; Tanimoto et al. 2010; Zhang et al. 2014; Soulier et al. 2018; Glöser et al. 2013; Elshkaki et al. 2016) from which the majority of tellurium is obtained where not included in this review, unless they focused specifically on the tellurium cycle as well. Similarly, only studies from geological surveys published in English language were included in the review although other geological surveys also provide information on the geological stocks of tellurium.

We selected sixteen studies following these criteria (Table S1):

Table S1. Literature dealing with anthropogenic tellurium cycles.

No.	Reference	Category[a]	Publisher[b]	Type	Spatial Boundary	Temporal Boundary	LC Stage(s)	Details
1	(Kavlak and Graedel 2013)	P	U	Dynamic	Global	1940-2010	All	MFA
2	(Zimmermann 2013)	R	U	Dynamic	EU	1990-2050	Installation, use, and (future) disposal of PV cells	MFA, CdTe PV modules
3	(Marwede and Reller 2012)	P	U	Dynamic	Global	2010-2040	Installation, use, and (future) disposal of PV cells	MFA, CdTe PV modules
4	(Fthenakis et al. 2009)	P	U	Static	Global	~2000	Ore-to-metal	LCA
5	(Fthenakis 2004)	P	U	Static	Global	~2000	Ore-to-metal	LCA
	(Classen et al. 2009)	R	O	Static	Global	~2000	Ore-to-metal	LCA
6	(Peiró et al. 2013)	P	U	Static	Global	~2008	Ore-to-tellurium-containing end products	MFA
7	(Graedel et al. 2015)	P	U	Static	Global	2008	Only end-use shares	Part of a criticality/substitutability assessment
8	(Nassar et al. 2012) (Yale criticality study)	P	U	Dynamic	Global and USA	2008	All	Streamlined MFA (depletion time model)
9	(EC 2017)EU criticality study	R	G	Static	EU-28	2017	All	Criticality study
10	(Bustamante and Gaustad 2014)	P	U	Dynamic	Global	2010-2060	Cu-Te byproduct system	MFA, CdTe PV
11	(Elshkaki and Graedel 2013)	P	U	Dynamic	Global (divided into world regions)	2010-2050	CdTe module	MFA

No.	Reference	Category[a]	Publisher[b]	Type	Spatial Boundary	Temporal Boundary	LC Stage(s)	Details
12	(Marwede and Reller 2014)	P	U	Static	Global	2010	CdTe module	MFA
13	(Calvo et al. 2016, 2018)	P	U	Static	EU-28	2014	All	MFA
14	(Wang et al. 2018)	P	U	Dynamic	Global	1930-2010	All	MFA
15	(USGS 2018, 2016)	R	G	Static	Global and USA	2015 - 2016	Refinery production	Geological Survey
16	(BGS 2017)	R	G	Static	Global	2012-2016	Refinery production	Geological Survey

[a] Journal Paper: P, Report: R, Other: O

[b] University: U, Industry: I, Government: G, NGO: Non-Governmental Organization

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